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## Aerosol radiative effects in photosynthetically active radiation and total irradiance at a Mediterranean site from an 11-year database

Lozano, Ismael López

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1 **Aerosol radiative effects in photosynthetically active radiation and total**  
2 **irradiance at a Mediterranean site from an 11-year database**

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11 **Abstract**

12 This study addresses the analysis of the aerosol radiative forcing (ARF) and aerosol  
13 forcing efficiency (AFE) at surface in the Photosynthetically Active and Total radiation  
14 ranges in a Southwest Mediterranean site. A thorough analysis of a long-term database  
15 (2008-2018) has been performed, bringing very valuable results about both, the  
16 absolute values and trends in ARF and AFE for both spectral intervals. The largest  
17 monthly mean for aerosol optical depth at 500 nm (AOD<sub>500</sub>) is found in summer (0.16 at  
18 July and August) meanwhile the lowest value is in winter (0.08 at November and  
19 December), with an interannual range varying from  $0.11 \pm 0.03$  (in 2018) to  $0.17 \pm 0.03$   
20 (in 2014). The AFE variation range has been estimated between  $-12$  and  $-198 \text{ Wm}^{-2}\tau^{-1}$  for  
21 PAR and between  $-9$  and  $-450 \text{ Wm}^{-2}\tau^{-1}$  for Total irradiance. ARF varies between  $-1 \text{ Wm}^{-2}$   
22 and  $-23 \text{ Wm}^{-2}$  in the PAR range, taking values from  $-1$  to  $-40 \text{ Wm}^{-2}$  in the Total one. This  
23 result points out the relevance of the aerosol effects on the PAR range, which can involve  
24 up to a 50% of the Total ARF. Moreover, a notable dependence of ARF and AFE on the  
25 solar position has been detected, increasing their absolute values at solar zenith angle

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26 from 0° to 45°-60° and decreasing to zero for lower solar positions. Additionally, this  
27 analysis has revealed the existence of a significant downward trend in AFE values for  
28 PAR, with a slope of  $2.7 \text{ Wm}^{-2}\tau^{-1}\text{year}^{-1}$ . Although the slope is positive, taking into account  
29 that the AFE values are negative, the slope value implies that the aerosol cooling  
30 radiative effect of aerosols is decreasing. However, no trends have been detected  
31 neither in AFE nor ARF values in the Total solar range. These results evidence the long-  
32 term aerosol effects over the different spectral intervals and emphasize the need for  
33 detailed analysis of the aerosol radiative effects on fundamental spectral intervals such  
34 as the PAR range.

35 Keywords: Aerosols; Photosynthetically active radiation; Radiative forcing.

36

## 37 **1.- INTRODUCTION**

38 Photosynthetically Active Radiation (PAR) is commonly defined as the electromagnetic  
39 radiation in the waveband between 400 and 700 nm (McCree, 1972). This spectral  
40 interval, which contains the maximum of the solar radiation spectrum, plays a  
41 fundamental role in vegetation productivity and agricultural research (Caya et al., 2018;  
42 McCree, 1981). PAR is the driver of the photosynthesis process and the biochemical  
43 reactions involved in it (Wu et al., 2019) and, therefore, the beginning of the plant  
44 growth. Moreover, PAR is a key factor controlling ecological processes such as the  
45 terrestrial carbon and hydrological cycles (Jonard et al., 2020; Potter et al., 2007, 2008).  
46 Along its path throughout the atmosphere, solar radiation, and particularly PAR, is  
47 attenuated by scattering and absorbing processes, being atmospheric aerosols, the main

48 factor determining the amount and distribution of solar radiation reaching the Earth's  
49 surface in absence of clouds. Aerosol particles affect the Earth's radiation budget both  
50 directly, by scattering and absorption, and indirectly, modifying cloud properties (e. g.  
51 Eswaran et al., 2019; Farahat et al., 2016; Satheesh & Krishna Moorthy, 2005). Aerosol  
52 attenuation presents an important spectral dependence. Thus, while spectral aerosol  
53 absorption decreases with wavelength, aerosol scattering efficiency strongly depends  
54 on the aerosol composition, increasing with wavelength for mineral dust and decreasing  
55 in case of urban pollution (Bergstrom et al., 2007). Due to this spectral dependence of  
56 the attenuation processes, aerosol effects over shorter wavelengths, such as the PAR  
57 interval, take a special relevance (Xu et al., 2003).

58 In order to quantify the radiative balance variations due to changes in atmospheric  
59 aerosols, the concepts of aerosol radiative forcing (ARF) and aerosol forcing efficiency  
60 (AFE) are widely employed. ARF is defined as the change in the net radiation due to  
61 variations in the atmospheric aerosol properties with respect to an aerosol-free  
62 atmosphere. ARF highly depends on the aerosol size distribution and composition (e.g.  
63 Foyo-Moreno et al., 2014). Thus, while mineral dust particles show negative ARF values  
64 associated to a strong cooling, the anthropogenic aerosols exhibit a complex behavior,  
65 with positive and negative ARF values depending on many factors such as greenhouse  
66 gases and surface changes (e.g. Andreae et al., 2005; Charlson et al., 1991; Esteve et al.,  
67 2012; Gopal et al., 2014; Hansen et al., 2011; Satheesh & Krishna Moorthy, 2005; Zhuang  
68 et al., 2013). On the other hand, AFE is defined as the rate at which the atmosphere is  
69 forced per unit of aerosol optical depth, allowing for a more detailed assessment of the  
70 radiative forcing considering the aerosol type.

71 Several studies have analyzed the impact of different aerosol types, and particularly  
72 their ARF, on total solar radiation (280-3000 nm) worldwide (e.g. Sicard et al., 2016;  
73 Sorribas et al., 2019; Zhang et al., 2018). However, aerosol radiative effects and its  
74 relationship to climate change remain inaccurate (IPCC 2013; Stocker et al., 2013). This  
75 uncertainty is larger in the PAR range because of the scarcity of related studies (Lyamani  
76 et al., 2006a; Mateos et al., 2014; Zhu et al., 2015). In fact, a worldwide routine network  
77 for the measurement of PAR is not yet established and PAR is often calculated as a  
78 constant ratio of the Total irradiance (Alados et al., 1996; Ge et al., 2011). Thus, one of  
79 the main drawbacks for this type of analysis is the lack of simultaneous and reliable  
80 measurements of PAR and aerosol properties. This limitation is stronger when the  
81 analysis is focused on trends in long-term databases and on the Mediterranean region  
82 (Di Biagio et al., 2009).

83 Recently, Obregón et al. (2020) studied the spatial and temporal AOD variations and the  
84 effects on solar radiation at the surface in the Mediterranean basin during a long period  
85 (2000–2018). Previously, they quantified ARF and AFE at Évora (Portugal, Southwestern  
86 Iberian Peninsula) during thirteen years (Obregón et al., 2017). In this Mediterranean  
87 region, temperature is increasing faster than the world average during the last decades  
88 (Lionello et al., 2014), and climate projections predict an increase of extreme climatic  
89 events, such as heat waves and droughts (Garcia-Herrera et al., 2014; Lionello et al.,  
90 2014). Besides, the Mediterranean region is subject to high aerosol loads, especially  
91 during spring and summer (Nabat et al., 2015), leading this region as a benchmark for  
92 climatic effect studies.

93 In this context, this aims to assess the aerosol radiative effects at surface on PAR for an  
94 urban middle-latitude site (Granada) in the Mediterranean basin for the decade 2008-  
95 2018. Local aerosols sources are traffic, local mineral dust during the dry season, and  
96 anthropogenic aerosols in winter from fuel oil combustion for domestic heating (Titos  
97 et al., 2012, 2017). At the same time, this site is also frequently influenced by emissions  
98 of several allochthonous aerosols sources such as continental aerosols from Europe and  
99 mineral dust from Africa (Fernández et al., 2019; Guerrero-Rascado et al., 2008, 2009;  
100 Lyamani et al., 2006a, 2006b, 2010), transported smoke from North America, North  
101 Africa and Europe (Alados-Arboledas et al., 2011; Baars et al., 2019; Ortiz-Amezcuca et  
102 al., 2014; 2017; Titos et al., 2017), extraordinarily, aerosols events from volcanic plumes  
103 (Navas-Guzmán et al., 2013; Sicard et al., 2012 ), and oceanic aerosols from Arctic and  
104 Atlantic oceans or maritime aerosols from the Mediterranean sea (Cariñanos et al.,  
105 2021; Pérez-Ramírez et al., 2016). Due to this variety in aerosol sources and types,  
106 aerosols over Granada are complex and variable making this an attractive region for the  
107 analysis of the aerosol radiative effects.

108 To this aim, AFE and ARF values for the PAR and Total ranges for cloud-free situations  
109 have been estimated, and analyzed in detail for different years and solar positions.  
110 Additionally, potential AFE and ARF trends at both spectral ranges, PAR and Total, have  
111 been assessed and compared.

112

## 113 **2.- EXPERIMENTAL SITE AND DATASET**

114 Measurements used in this study have been collected at the radiometric station  
115 installed on the roof of the IISTA-CEAMA building at Granada (37.16 °N, 3.61 °W, 680  
116 m.a.s.l.), an urban site located in the Southeast of Spain in the West Mediterranean  
117 region. This radiometric station is managed by the Atmospheric Physic Research Group  
118 (GFAT) at the University of Granada and is part of the observatory AGORA (Andalusian  
119 Global ObservatoRy of the Atmosphere) in the framework of ACTRIS (Aerosol, Clouds  
120 and Trace Gases Research Infrastructure). The period analyzed encompasses the decade  
121 2008-2018. Granada presents large seasonal temperature differences, characterized by  
122 cool winters and hot summers, with mean daily maximum temperature at surface of  
123  $(14.6 \pm 2.4)$  °C in winter and  $(32 \pm 3)$  °C for 1981-2010 period (AEMET, Spanish  
124 Meteorology Statal Agency).

125 In this study, one-minute measurements of Photosynthetically Active Radiation (PAR)  
126 were measured with a SKP 215 PAR Quantum Sensor (#28715) manufactured by Skye  
127 Instruments. This instrument measures the solar radiation in the range of 400-700 nm  
128 using a blue enhanced planar diffused silicon detector with a sensitivity of 0.015  
129  $\mu\text{A}/\mu\text{mol m}^{-2}\text{s}^{-1}$ . The quantum sensor has a maximum relative error <5%. Simultaneous  
130 one-minute measurements of total solar irradiance were recorded with a radiometer  
131 CM11 (#861452) manufactured by Kipp&Zonen. This instrument measures broadband  
132 solar irradiance in the range of 280-2800 nm and complies with International  
133 Organization for Standardization (ISO) 9060 criteria for an ISO secondary standard  
134 pyranometer. Both measurements were recorded in a CR10X data logger manufactured  
135 by Campbell Scientific.

136 Additionally, Aerosol Optical Depth (AOD) values, measured every 15 minutes by a  
137 CIMEL Sun/sky photometer (CE-318-4) were used in this study. This instrument, which  
138 is integrated in the AERONET network (Holben et al., 1998), measures direct solar  
139 irradiance with a 1.2° full field of view at 340, 380, 440, 500, 675, 870, 940 and 1020 nm  
140 as well as sky radiances in the almucantar and principal plane geometries at 440, 675,  
141 870 and 1020 nm. All radiance measurements are processed following the AERONET  
142 protocol as described by Holben et al. (1998), obtaining columnar aerosol properties at  
143 different quality levels (1.0, 1.5, 2.0). Version 3 of AOD at level 2.0 (Giles et al., 2019),  
144 the highest quality AERONET data, was used in this study, except for 2014 for which only  
145 version 2 at 2.0 level AOD was available. AOD data has a total uncertainty of 0.01 for  
146 wavelengths  $\geq 440$  nm and 0.02 for shorter wavelengths (Holben et al., 1998). Sun-  
147 photometer also provided the surface albedo measurements used in this study at 440,  
148 675, 870 and 1020 nm with a total uncertainty of 0.02 (Foyo-Moreno et al., 2014). Both,  
149 CIMEL photometer and radiometers involved in this study have been intercompared  
150 respect to reference instruments several times along the 11-year period analyzed, with  
151 their last intercomparison dated on May 2020 for the CIMEL photometer, May 2019 for  
152 the CM11 pyranometer and August 2020 for the PAR sensor. CIMEL were calibrated  
153 following AERONET protocols (Holben et al., 1998) at the RIMA calibration facilities at  
154 Valladolid, Spain, while the radiometers have been intercompared with a Kipp&Zonen  
155 CMP21 and a LICOR-190SA, respectively, following WMO procedure for intercomparison  
156 (WMO, 2008). Particularly, the calibration factors applied in this study showed a change  
157 of  $0.15 \text{ mV/Wm}^{-2}$ , in CM11 pyranometer, and  $0.4 \text{ mV/Wm}^{-2}$ , in PAR sensor, for the entire  
158 period 2008-2018. This involves an annual average change of  $0.013 \text{ mV/Wm}^{-2} \text{ year}^{-1}$  and  
159  $0.027 \text{ mV/Wm}^{-2} \text{ year}^{-1}$  for Total and PAR irradiance sensor, respectively. Both values are



160 notably below the maximum change per year (long-term stability) detailed by the  
161 corresponding manufacturer, 0.5% for the CM11 and 2% for SKP 215 PAR Quantum  
162 Sensor, ensuring the calibration factor stability required for a trend analysis.

163

### 164 **3.- METHODOLOGY**

#### 165 **3.1 Data analysis and selection of cloud-free scenarios**

166 To perform the analysis of the aerosol radiative effects, a database of simultaneous  
167 measurements of PAR, Total irradiance and AOD has been built for the period 2008-  
168 2018. This vast database ensures a large variety of seasonal processes, solar geometries  
169 and meteorological conditions and guarantees the representativeness of the dataset for  
170 the analysis proposed here.

171 A deep quality control has been applied to this final dataset in order to detect and  
172 eliminate potential erroneous measurements. First, only those measurements recorded  
173 for solar zenith angles smaller than  $80^\circ$  have been selected to avoid solar radiation data  
174 affected by a cosine response error, the maximum difference from the ideal response  
175 for PAR Quantum Sensor was approximately 7% at a zenith angle of  $80^\circ$  (Akitsu et al.,  
176 2017). Additionally, those cases in which total global irradiance reached higher values  
177 than extraterrestrial total irradiance ( $k_t > 1$ ) or diffuse irradiance higher than global  
178 irradiance ( $k_d > 1$ ) were removed. Outliers were detected by visual inspection and  
179 consequently removed. Possible troubles associated with power supply and  
180 temperature of the acquisition system were also checked.

181 Additionally, a thorough analysis of the AOD values along the period of study has been  
182 performed. Thus, data has been monthly grouped along the entire period in order to  
183 analyze seasonal evolution and a detailed statistical has been computed including  
184 arithmetic mean (Ave), standard deviation (SD), median (Md), minimum (Min),  
185 maximum (Max), percentiles 5<sup>th</sup>, 25<sup>th</sup>, 75<sup>th</sup> and 95<sup>th</sup> (P5, P25, P75 and P95, respectively),  
186 skewness (Ske), kurtosis (Kur) and the variation coefficient (VC) defined as the ratio  
187 between SD and the arithmetic mean.

188 In order to only account for aerosol effects, cloud-free situations have been selected  
189 applying the criterion proposed by Alados-Arboledas et al. (2000). This criterion detects  
190 as cloud-free situations those in which:

$$191 \quad k_t > 0.53 + 0.31 \cos \text{SZA} - 0.15 \cos^2 \text{SZA} \quad (1)$$

192 where  $k_t$  is the clearness index, defined as the ratio between the total irradiance ground-  
193 measured and the extraterrestrial total irradiance ( $G_{\text{ext}} = E_o I_{\text{sc}} \cos \text{SZA}$ ), both on a  
194 horizontal plane, where SZA is the solar zenith angle,  $E_o$  is the eccentricity correction  
195 factor and the value used of the Solar Constant ( $I_{\text{sc}}$ ) is  $1367 \text{ Wm}^{-2}$  (Iqbal, 1983). The  
196 variability of the Solar Constant on the 11-year solar cycle has not been considered in  
197 this study. This empirical criterion, based on coefficients derived from a fitting to a  
198 polynomial function has been explicitly developed for Granada. This criterion only  
199 requires global irradiance measurements, commonly available at most radiometric  
200 stations.

### 201 **3.2 AFE and ARF estimation**

202 The ARF is defined as the difference between the measured net irradiance ( $F_{net}$ ) and the  
203 same magnitude for an aerosol-free atmosphere ( $F_{net,a}$ ):

$$204 \quad ARF = F_{net} - F_{net,a} \quad (2)$$

205 where  $F_{net}$  is the difference between the downward and the upward irradiances at the  
206 Earth's surface. Net irradiances under the presence/absence of aerosols can be written,  
207 respectively, as:

$$208 \quad F_{net} = (1 - A)I \quad (3)$$

$$209 \quad F_{net,a} = (1 - A)I_a \quad (4)$$

210 where  $I$  is the experimental irradiance,  $I_a$  is the estimated irradiance under absence of  
211 aerosols, and  $A$  is the surface albedo.

212 The aerosol forcing efficiency (AFE) is defined as the change in ARF per unit increase in  
213 AOD for a certain wavelength (Bush and Valero, 2003):

$$214 \quad AFE = \frac{dARF}{dAOD} \quad (5)$$

215 Thus, AFE at surface can be computed from the slope of the linear regression between  
216 ARF and AOD at fixed SZA (Antón et al., 2011; Díaz et al., 2007; García et al., 2006).

217 Different methodologies to estimate ARF have been proposed in literature. An extended  
218 method involves the use of radiative transfer models to estimate the net irradiance  
219 fluxes (Eswaran et al., 2019; Mateos et al., 2014; Sivan & Manoj, 2019). However, this  
220 procedure involves relevant assumptions such as the atmospheric composition and the  
221 aerosol layer description, which could lead to important errors. In this study, the so-

222 called direct method proposed by Satheesh and Ramanathan (2000) has been used.  
223 Once cloud-free situations have been selected from the database, AFE is derived as the  
224 slope of the linear fit between the experimental  $F_{net}$  values and AOD at fixed SZA:

$$225 \quad AFE = \frac{dF_{net}}{dAOD} \quad (6)$$

226 Then, ARF at surface is obtained as a result of multiplying AFE by the annual AOD average  
227 at the corresponding solar position (Di Biagio et al., 2010; Foyo-Moreno et al., 2014).  
228 The advantage of this method is that AFE is directly computed from the experimental  
229 data without further assumptions on the radiative fluxes under aerosol-free conditions.  
230 This method shows an important dependence on the solar zenith angle and, therefore,  
231 its application and analysis is limited to specific solar zenith intervals.

232 Particularly, AOD values at 500 nm have been used to analyze the aerosol effects on the  
233 PAR for roughly being the central wavelength in this spectral range. On the other hand,  
234 AOD at 675 nm has been chosen to estimate AFE and ARF for the Total irradiance  
235 considering that the central wavelength of the solar spectrum is roughly 680 nm (Di  
236 Biagio et al., 2010). This decision is also supported by previous works (e.g. Foyo-Moreno  
237 et al., 2014; Li et al., 2020; Romano et al., 2016).

238 Additionally, five categories of solar zenith angles to compute both AFE and ARF, namely  
239  $15^\circ$ ,  $30^\circ$ ,  $45^\circ$ ,  $60^\circ$  and  $75^\circ (\pm 1^\circ)$ , have been considered, in order to cover the majority of  
240 solar positions. Moreover, surface albedo provided by AERONET at 675 nm for our  
241 station has been used. Annual average surface albedo was estimated and employed in  
242 the calculation of the ARF, being 0.14 the annual average for all years, except for 2009

243 and 2013 for which a value of 0.15 was found. The overall error in next flux increases by  
244 less than 0.3% due to the uncertainty in surface albedo (Di Biagio et al., 2010).

### 245 **3.3 Trend analysis**

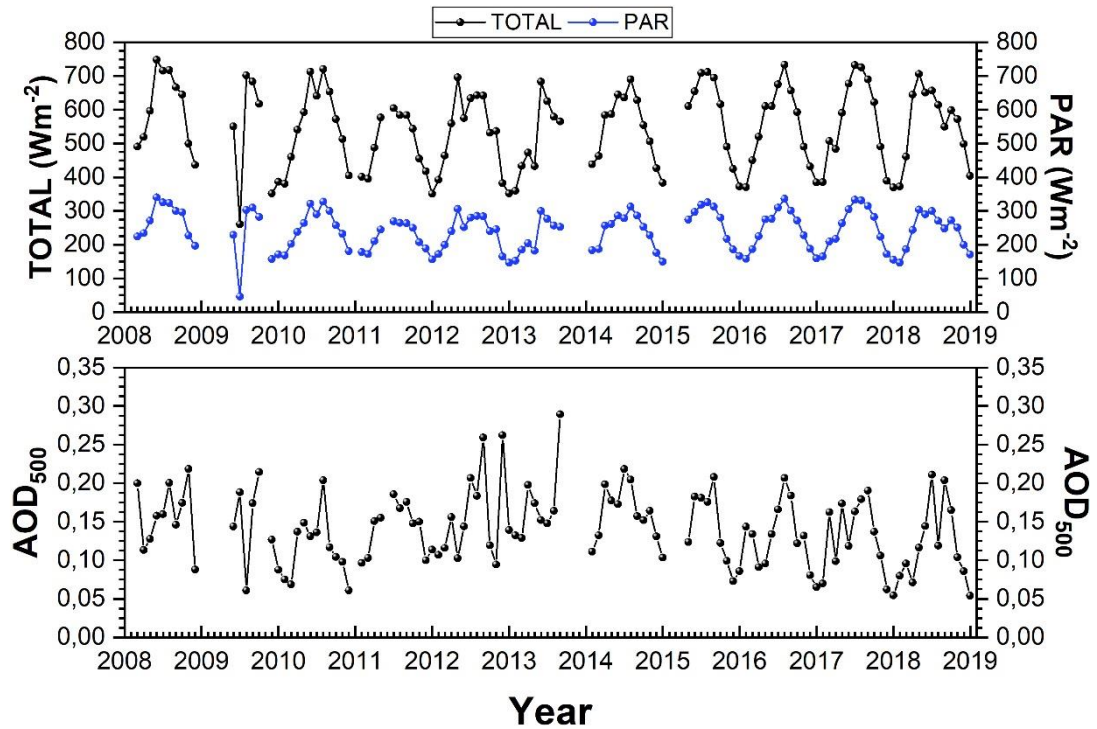
246 Finally, the non-parametric Mann-Kendall test (Mann, 1945) has been applied to detect  
247 time series trends for AOD, ARF and AFE with statistical significance. In addition, the Sen  
248 estimation of the trend slope has been performed, which complements the Mann-  
249 Kendall test (Sen, 1968). The use of the Sen method is appropriate for evaluating trends  
250 in time series as it is not affected by outliers and gaps, making it a common method in  
251 literature (e.g. Buffoni et al., 1999; Da Silva et al., 2010; Dadashi-Roudbari and Ahmadi,  
252 2020; Kodera et al., 2008; Kuo et al., 2020; Olmo & Alados-Arboledas, 1995; Zou et al.,  
253 2016). To perform these calculations the kbttau.m software developed by Jeff Burkey  
254 (*Mann-Kendall Tau-b with Sen's Method (Enhanced)*, 2020) was used.

255

## 256 **4.- RESULTS AND DISCUSSION**

### 257 **4.1 AOD characterization**

258 First, to have a brief global view, a general characterization of the whole databases  
259 including all variables analysed has been included in Figure 1, showing the monthly  
260 averages for every year. The two data series for the solar irradiance measurements  
261 display the same typical annual cycle with summer maximum (for example,  $(730 \pm 60)$   
262  $\text{Wm}^{-2}$  for Total and  $(340 \pm 30) \text{Wm}^{-2}$  for PAR in July 2016) and winter minimum, driven  
263 by the annual course of the solar zenith angle, with interannual variability.



264

265 Figure 1. Time series of monthly mean PAR (black line), Total (blue line) and AOD at  $\lambda=500\text{nm}$   
 266 ( $\text{AOD}_{500}$ ), for 2008-2018 years.

267

268 Focusing on AOD, for the sake of clarity, the analysis of AOD is presented here based on  
 269 500 nm (Table 1 and Figure 2). The largest monthly mean  $\text{AOD}_{500}$  is found in summer  
 270 (0.16 at July and August) meanwhile the lowest values are in winter (0.08 at November  
 271 and December. The maximum/minimum values occur, respectively, in summertime due  
 272 to the higher frequency Saharan dust outbreaks (Salvador et al., 2014), and in  
 273 wintertime due to the low probability of these events (Gkikas et al., 2013, 2018; Querol  
 274 et al., 2009) over the Mediterranean region. This behaviour agrees with previous  
 275 studies. Mateos et al. (2014) reported a minimum and maximum  $\text{AOD}_{440}$  in November  
 276 and July, respectively, at the same location during the period 2004-2012. Ant3n et al.  
 277 (2011) found minimum  $\text{AOD}_{380}$  ( $0.14 \pm 0.05$ ) and  $\text{AOD}_{400}$  ( $0.12 \pm 0.05$ ) in November and

278 December, respectively, coinciding in time with our minimum AODs, but differing with  
 279 respect to maximum values found in May ( $0.26 \pm 0.12$  for AOD<sub>380</sub> and  $0.24 \pm 0.12$  for  
 280 AOD<sub>440</sub>). These differences might be caused by the short period used (2006-2008),  
 281 probably limiting its representativeness. In a later study, Mateos et al. (2015) studied  
 282 the aerosol load over the Iberian Peninsula in five geographical sectors and they found  
 283 values for AOD<sub>440</sub> varying between 0.15 and 0.20. This annual cycle is also found by  
 284 Bennouna et al. (2016) for long measurement records (2003-2014) obtained at two sites  
 285 of Spain located in the North Central region, with values for AOD<sub>440</sub> of  $0.16 \pm 0.09$  in June  
 286 and  $0.08 \pm 0.06$  in December. Segura et al. (2017) found a mean value of  $0.15 \pm 0.11$  for  
 287 AOD<sub>550</sub> with the same seasonal pattern for a site located Mediterranean coastal area in  
 288 Spain during the period 2007-2016. Sicard et al. (2016) also found for AOD<sub>440</sub> a clear  
 289 annual cycle (maxima of 0.22 at Ersa and 0.27 at Palma observed in July) for a 5-year  
 290 period for different locations in the Western Mediterranean Basin.

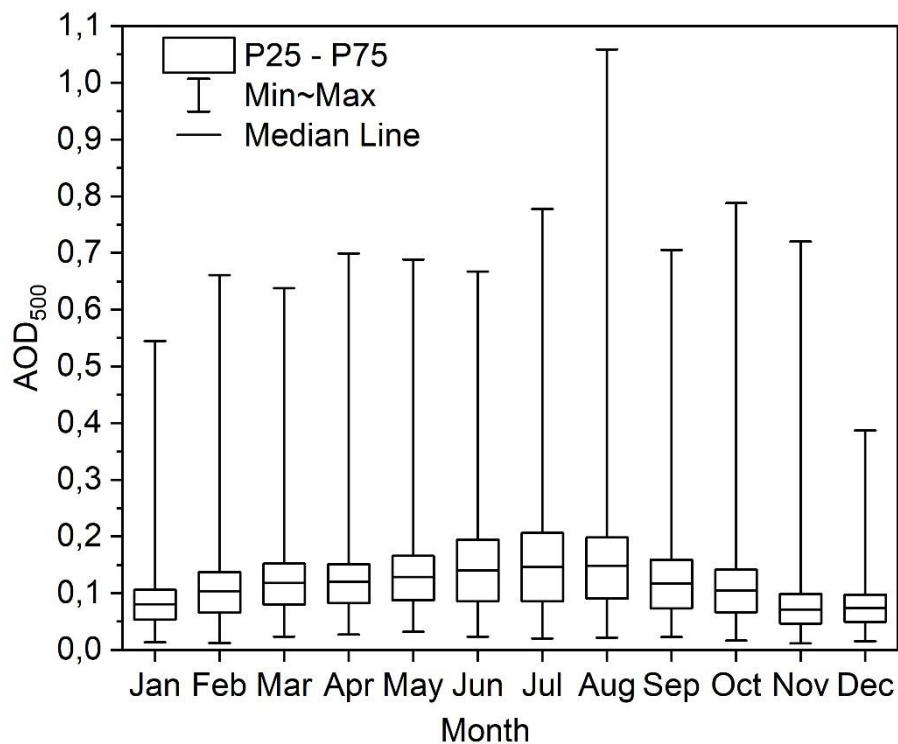
291

292 Table 1. Monthly statistics of AOD<sub>500</sub> for the period 2008-2018 based on daily values: Number  
 293 of datapoints (N), Average (Ave), Standard deviation (SD), Median (Md), Percentiles (P5, P25,  
 294 P75, P95), Skewness (Ske), Kurtosis (Kur) and coefficient of variation (CV).

Month	N	Ave	Md	P5	P25	P75	P95	Ske	kur	CV(%)
January	3603	0.09±0.06	0.08	0.03	0.06	0.11	0.20	2.19	10.92	63
February	3840	0.11±0.06	0.10	0.04	0.07	0.14	0.21	1.73	9.51	54
March	4641	0.13±0.07	0.11	0.05	0.08	0.16	0.26	1.89	9.20	54
April	5158	0.13±0.07	0.12	0.05	0.08	0.16	0.25	2.20	11.36	54
May	6986	0.13±0.07	0.13	0.06	0.09	0.17	0.25	2.17	12.12	51
June	10344	0.15±0.09	0.14	0.05	0.09	0.21	0.34	1.50	5.65	60
July	13132	0.16±0.09	0.15	0.05	0.09	0.22	0.34	1.20	4.61	58
August	10763	0.16±0.09	0.15	0.05	0.10	0.22	0.34	1.39	6.73	58
September	8447	0.13±0.07	0.12	0.05	0.08	0.17	0.26	1.35	5.43	55
October	6529	0.11±0.06	0.10	0.04	0.07	0.15	0.24	1.51	7.57	56
November	4771	0.08±0.05	0.07	0.03	0.05	0.10	0.16	2.90	20.63	63
December	3436	0.08±0.04	0.07	0.03	0.05	0.10	0.16	1.68	7.67	53

295

296 The analysis of P5 and P95 is similar to the trend of the average values. The median AOD  
297 values are smaller than the mean, what is a common feature over the Iberian regions  
298 (Mateos et al., 2015). The absolute difference between the median and third quartile is  
299 also larger than the absolute difference between the median and the first quartile  
300 except at two months, namely April and May. This last result is also in accordance with  
301 those found by Mateos et al. (2015). The kurtosis and asymmetry data for AOD show  
302 that the distribution for all months is leptokurtic and positive asymmetric, obtaining the  
303 highest asymmetry values for November, while the lowest values are found in July.



304

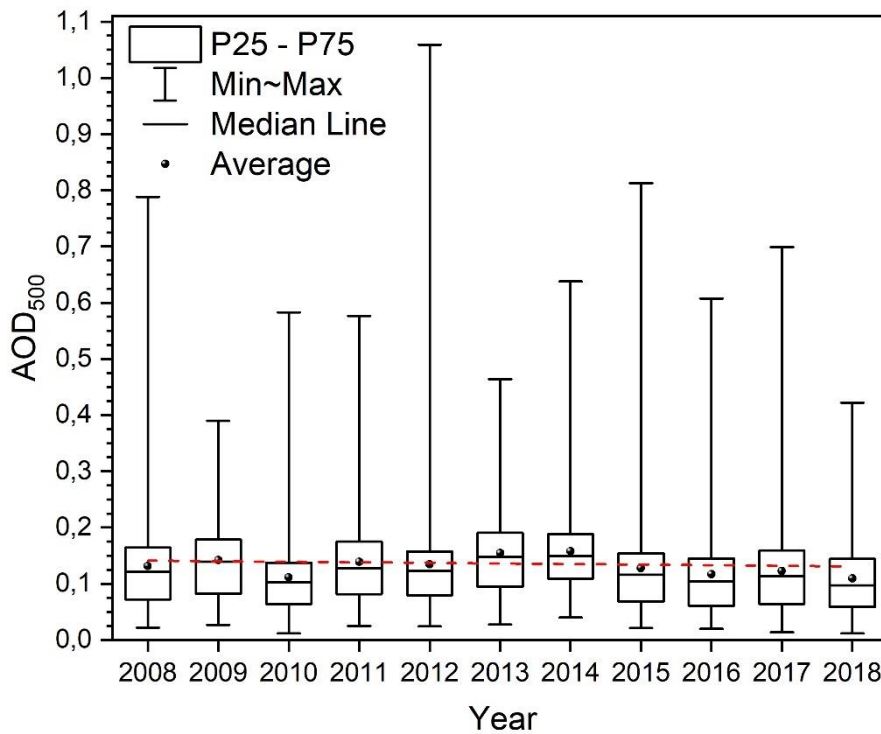
305 Figure 2. Monthly statistics of AOD<sub>500</sub> for the period 2008-2018. Bars correspond to the  
306 minimum and maximum values, the box limits are the P25 and P75 percentiles and the midline  
307 is the median.



308 The box-whisker diagram plot in Figure 2 showed a clear seasonality (already observed  
309 in Table 1) with higher values in central months, with a high standard deviation specially  
310 at August due to African dust intrusions additionally to resuspension processes of local  
311 mineral aerosols owing to the dryness of the soil. Thus, the maximum AODs were found  
312 in summer and minimum values in winter. This evident AOD annual pattern is also found  
313 at seven sites in the Iberian Peninsula covering different aerosol types and  
314 environmental conditions during three coincident years (2010-2012) (Foyo-Moreno et  
315 al., 2019). The interquartile range P75-P25 is also larger in summer. It is worthy to note  
316 relatively high values of maximum AOD<sub>500</sub> (0.66) and its P95 (0.21) in February. Despite  
317 of the low frequency of Saharan dust events over the Mediterranean region in winter  
318 (Gkikas et al., 2013, 2018; Querol et al., 2009), this is explained due to the intense  
319 Saharan dust events occurring in February 2016 and 2017 (Cazorla et al., 2017;  
320 Fernández et al., 2019), and also the increase in the anthropogenic local emissions in  
321 winter in addition to the orographic and meteorological conditions of Granada which  
322 favors the particle stagnation (Lyamani et al., 2012).

323 To study the interannual variability the box-wisher diagram plot of AOD<sub>500</sub> at Granada is  
324 shown in Figure 3, for 2008-2018 years. The interannual range of AOD<sub>500</sub> varies from  
325  $0.11 \pm 0.08$  in 2018 to  $0.16 \pm 0.14$  in 2014 for, and showed firstly a decreasing trend in  
326 the subperiod 2008-2010, an increasing trend in 2010-2014 and a latter decreasing trend  
327 from 2014, but the Mann Kendall test revealed a slope of -0.001 with a p-value of 0.53  
328 and, therefore, with no trend for AOD<sub>500</sub>. The years with higher variability are 2012, 2015  
329 and 2016, with a maximum difference between the extreme values for 2012. In general,  
330 a clear decrease in the aerosol load over the Iberian Peninsula has been observed since

331 the 2000s. In particular, Mateos et al., 2015 found a decrease of -0.07 per decade in  
 332 AOD<sub>400</sub> for the Southeastern sector. Li et al. (2014) found a decreasing trend for a large  
 333 stations number around the world (including Granada). Thus, the largest decreases were  
 334 found over western Europe, reaching -0.1 per decade, and particularly they found at  
 335 Granada a slope of -0.03 per decade for AOD<sub>400</sub>. In our study, no significant trend has  
 336 been found for AOD<sub>500</sub> in the whole data.

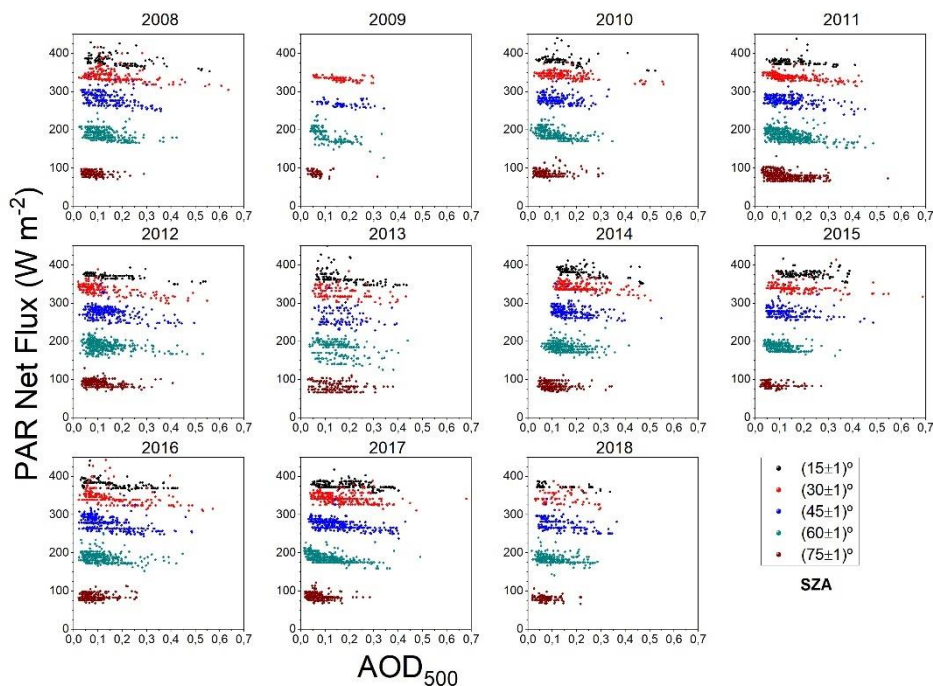


337

338 Figure 3. Annual statistics of AOD<sub>500</sub> for the period 2008-2018. Bars correspond to the  
 339 minimum and maximum values, the box limits are the P25 and P75 percentiles, the midline is  
 340 the median and the sphere is the mean value. Dashed lines point out the linear trend  
 341 evaluated by the Sen method.

## 342 4.2 Analysis of AFE and ARF

343 Figure 4 shows the relationship between PAR and AOD<sub>500</sub> for the entire studied period  
 344 at different solar zenith angles centered at 15°, 30°, 45°, 60° and 75° ( $\pm 1^\circ$ ). For each  
 345 year, five well-differentiated point clouds are observed, corresponding each of them to  
 346 one of the five solar positions analyzed. As it was expected, the  $F_{\text{net}}^{\text{PAR}}$  increases for  
 347 lower solar zenith angles due to the lower solar radiation path through the atmosphere  
 348 at this solar position, increasing the solar irradiance reaching the surface. Additionally,  
 349 a larger spread of the datapoints is observed for low values of AOD because of the large  
 350 influence of the measurement uncertainty on the low AOD values. A similar behaviour  
 351 has been observed for  $AFE^{\text{total}}$  (not shown here). An additional explanation of this  
 352 datapoint large spread can be the existence of numerous sources of aerosols, which  
 353 gives this region a high variability and complexity (Benavent-Oltra et al., 2017; Bravo-  
 354 Aranda et al., 2015; Cazorla et al., 2017; Córdoba-Jabonero et al., 2011; Pérez-Ramírez  
 355 et al., 2016).



356

357

Figure 4. Evolution of PAR Net Flux versus AOD<sub>500</sub> for the period 2008-2018.

358

359 As it was described in Section 3.2, the slope of the linear fit at each angle category in

360 Figure 4 represents the AFE. Both, AFE<sup>PAR</sup> and AFE<sup>total</sup> present negative values (Table 2),

361 leading to a cooling effect by aerosols over Granada (Granados-Muñoz et al., 2019).

362 AFE<sup>PAR</sup> varies between -12 and -198 Wm<sup>-2</sup>τ<sup>-1</sup> while AFE<sup>total</sup> ranges between -9 and -450363 Wm<sup>-2</sup>τ<sup>-1</sup>, showing a higher variability than in the PAR interval. These results agree with364 previous studies. Thus, Di Biagio et al. (2010) accounted for an AFE<sup>total</sup> value of -309 ± 16365 Wm<sup>-2</sup>τ<sup>-1</sup> for solar zenith angles between 15° and 25° and mixed aerosols at Lampedusa

366 (Italy). Lower values have been estimated for other aerosol types as desert mineral dust

367 and urban/industrial and biomass burning aerosols (Di Biagio et al., 2010). In this study,

368 AFE<sup>PAR</sup> entails between 20 and 60 % of the AFE<sup>total</sup> with an average value of 30% for the

369 whole dataset, which points out the relevance of the aerosol effects on PAR and the

370 influence of this spectral interval.

371

372 Table 2. Surface aerosol forcing efficiency (Wm<sup>-2</sup>τ<sup>-1</sup>) for PAR and Total irradiance (ARE<sup>PAR</sup> and373 ARE<sup>total</sup>, respectively) with its variability at one standard deviation level by years for different

374 solar zenith angles (SZA).

Year	AFE <sup>PAR</sup> (Wm <sup>-2</sup> τ <sup>-1</sup> )				
	SZA (°)				
	15	30	45	60	75
2008	-65±12	-71±10	-103±14	-83±11	-28±11
2009		-47±8	-37±13	-198±18	-35±23
2010	-49±15	-52±5	-52±15	-117±12	1±11
2011	-27±13	-55±6	-75±8	-101±7	-49±6
2012	-43±4	-76±7	-80±10	-51±8	-36±8
2013	-92±15	-76±17	-50±30	-90±20	-27±13
2014	-78±10	-49±7	-69±12	-40±10	-17±9
2015	-18±9	-33±8	-53±11	-85±10	-12±9
2016	-64±8	-82±11	-94±9	-34±8	32±8

2017	-27±5	-40±6	-74±5	-96±5	-40±8
2018	-42±1	-40±30	-83±13	-56±10	-24±14
$AFE^{total} (Wm^{-2}\tau^{-1})$					
SZA (°)					
Year	15	30	45	60	75
2008	-170±30	-183±23	-300±30	-220±30	-30±40
2009		-220±30	-155±4	-450±50	-90±60
2010	-150±37	-144±17	-210±40	-330±40	-70±30
2011	-192±34	-168±18	-199±22	-281±19	-116±16
2012	-114±13	-163±18	-180±30	-123±19	-70±20
2013	-270±50	-200±50	-120±70	-220±60	-60±40
2014	-128±22	-126±23	-210±40	-210±30	-9±24
2015	-84±23	-92±18	-150±30	-310±30	-50±30
2016	-203±21	-200±30	-240±21	-205±23	59±21
2017	-149±13	-156±15	-219±12	-374±15	-170±30
2018	-170±19	-190±70	-310±40	-400±40	-120±60

375

376 Regarding the AFE dependence on solar zenith angle, both  $AFE^{PAR}$  and  $AFE^{total}$  showed  
377 enhanced values at roughly 45° or 60°. Other authors have reported a different pattern  
378 of this dependence with relatively constant or opposite trend for lower values of SZA  
379 and the same decreasing trend obtained in this work for high SZA values. Thus, Di Biagio  
380 et al. (2009) pointed out that this trend depends on the aerosol type and seems to be  
381 reversed for urban/industrial-biomass burning aerosols. Additionally, several authors  
382 suggested that the inflection point in this trend depends on the aerosol properties (Di  
383 Biagio et al., 2009, 2010; Formenti et al., 2002). This dependence of AFE on SZA can be  
384 explained by the combination of different factors. As the solar radiation path increases  
385 in the atmosphere, the attenuation as well as the diffuse fraction increases, especially  
386 at shorter wavelengths. On the other hand, for high SZA, the atmosphere is optically  
387 thicker and the AFE tends to decrease. Consequently, the AFE displays a dependence on  
388 SZA, which confirms the need to estimate forcing efficiency at fixed solar position  
389 applying the direct method employed in this study (Di Biagio et al., 2009, 2010; Formenti  
390 et al., 2002; Meloni et al., 2005; Nemesure et al., 1995).

391 The same analysis has been carried out for the ARF. Table 3 shows the values for  $ARF^{PAR}$   
 392 and  $ARF^{total}$  during the full period, ranging from -1 to -23  $Wm^{-2}$  and from -1 to -40  $Wm^{-2}$   
 393 for  $ARF^{PAR}$  and  $ARF^{total}$ , respectively. These values are in accordance to those reported  
 394 by other authors. Thus, Meloni et al. (2005) found  $ARF^{PAR}$  values between -10 and -20  
 395  $Wm^{-2}$  at Lampedusa. Our work has reported a percentual ratio  $ARF^{PAR}/ARF^{total}$  of 50% in  
 396 average, that is a higher value than the one found for AFE (30%). This average  
 397 percentage found for ARF is higher than the mean value obtained at Granada for the  
 398 ratio PAR to Total (43%) with values varying between 33 and 52% (Foyo-Moreno et al.,  
 399 2017), highlighting the important role of the aerosols on PAR, greater than on Total.  
 400 Following Ma et al. (2007), the ratio PAR to Total irradiance for various locations around  
 401 the world present values between 35 and 58%.

402  
 403 Table 3. Surface aerosol radiative forcing ( $Wm^{-2}$ ) for PAR and Total irradiance ( $ARF^{PAR}$  and  
 404  $ARF^{total}$ , respectively) with its variability at one standard deviation level by years for different  
 405 solar zenith angles (SZA).

$ARF^{PAR}(Wm^{-2})$					
SZA ( $^{\circ}$ )					
Year	15	30	45	60	75
2008	-11±9	-12±9	-15±10	-10±7	-2±2
2009	-	-8±4	-6±5	-23±14	-3±3
2010	-7±7	-8±6	-6±5	-13±8	0±1
2011	-5±4	-9±6	-10±7	-13±9	-6±4
2012	-6±5	-12±10	-11±8	-6±5	-4±3
2013	-15±11	-12±9	-9±8	-13±9	-4±4
2014	-17±8	-10±5	-12±6	-6±4	-2±2
2015	-4±3	-6±5	-7±6	-9±5	-1±1
2016	-11±8	-14±11	-12±9	-3±3	3±2
2017	-4±3	-7±4	-11±7	-10±7	-3±2
2018	-6±4	-5±6	-11±8	-6±4	-2±2
$ARF^{total}(Wm^{-2})$					
SZA ( $^{\circ}$ )					
Year	15	30	45	60	75
2008	-21±19	-23±20	-31±25	-19±14	-2±3
2009	-	-26±13	-20±9	-40±30	-4±5

2010	-15±17	-15±15	-18±14	-25±18	-4±4
2011	-24±20	-21±17	-20±17	-26±20	-9±7
2012	-14±14	-22±21	-19±17	-11±10	-5±5
2013	-30±30	-25±22	-15±17	-24±20	-6±7
2014	-21±13	-20±13	-27±16	-23±13	-1±2
2015	-12±10	-12±13	-14±13	-21±14	-2±3
2016	-28±24	-30±30	-24±22	-15±13	3±4
2017	-18±13	-20±14	-24±17	-27±22	-7±5
2018	-16±14	-18±19	-30±24	-30±30	-5±6

406

407 Regarding the dependence of ARF on solar position, a similar pattern to that of AFE is  
 408 found. The absolute ARF values increase with increasing SZA up to 45° or 60°, depending  
 409 on the year considered, and then decrease. However, this dependence is less  
 410 pronounced than for AFE, with values relatively constant for SZA below an intermediate  
 411 value. The value of SZA for the change always is coinciding with the values obtained for  
 412 AFE and also is coinciding for PAR and Total irradiance except for 2018. This trend is  
 413 similar to that obtained by Meloni et al. (2005), but the trend is reversed for aerosols  
 414 more absorbing. This fact may explain the different behavior below a given SZA value.  
 415 Our work covers a long period including a wide variety of aerosols and, thus, this mixture  
 416 generally exhibits large values of single scattering albedo. In fact, the maximum values  
 417 of single scattering albedo registered at Granada during the years 2010-2012 are close  
 418 to 1, which indicates that Granada recorded events of non-absorbing aerosols (Foyo-  
 419 Moreno et al., 2019).

420 Comparing our results with previous studies, we found similar values of both AFE and  
 421 ARF for both PAR and Total irradiance. Antón et al. (2012) performed ARF and AFE  
 422 calculations at Granada for Total irradiance with another method and they found, values  
 423 of ARF between -100 and -200 Wm<sup>-2</sup> and AFE of -115 Wm<sup>-2</sup>τ<sup>-1</sup> at 675 nm during an African  
 424 dust event. For a longer period of time (2005-2010), Valenzuela et al. (2012) computed  
 425 values of ARF and AFE with the radiative transfer model SBDART during African dust

426 events, and found ARF from  $-13$  to  $-34 \text{ Wm}^{-2}$  and AFE from  $-65$  to  $-74 \text{ Wm}^{-2}\tau^{-1}$  at  $440 \text{ nm}$ ,  
427 depending on the mineral sources. Foyo-Moreno et al. (2014), using the direct method  
428 for the radiative effects calculations, found an ARF of  $-28 \text{ Wm}^{-2}$  and an AFE of  $-73.4 \text{ Wm}^{-2}\tau^{-1}$   
429  $\tau^{-1}$  at solar fixed angle of  $15^\circ$  during the period 2006-2007. Focusing on PAR Lyamani et  
430 al. (2006b) found values of ARF of  $-20.4 \text{ Wm}^{-2}\tau^{-1}$  during an African dust event in 2003  
431 and  $-16.1 \text{ Wm}^{-2}$  during intrusions from the Central Europe region, with AFE values of  $-$   
432  $73.4$  and  $-78.2 \text{ Wm}^{-2}\tau^{-1}$  at  $670 \text{ nm}$ . Therefore, all these studies, performed in the same  
433 area of study, found values that are within the ranges of our findings. Other authors  
434 focused on other regions of the Mediterranean basin. Meloni et al. (2005), using a  
435 radiative transfer model at  $400\text{-}700 \text{ nm}$ , found ARF daily mean values  $-12.9$  and  $-19.5$   
436  $\text{Wm}^{-2}$  in July and AFE at  $500 \text{ nm}$  ranging between  $-28.4$  and  $-30.1 \text{ Wm}^{-2}\tau^{-1}$  and between  
437  $-42.9$  and  $-45.6 \text{ Wm}^{-2}\tau^{-1}$  for several days at Lampedusa. Sicard et al. (2016) for  $50^\circ < \text{SZA}$   
438  $< 60^\circ$  found values of  $\text{ARF}^{\text{total}}$  of  $(-23 \pm 13) \text{ Wm}^{-2}$  and  $(-136 \pm 41) \text{ Wm}^{-2}\tau^{-1}$  for  $\text{AFE}^{\text{total}}$  in  
439 summer at Palma de Mallorca (Mallorca Island, Spain). All of these values are in good  
440 agreement with our findings.

441 However, it is necessary to emphasize the different methodologies used in the works of  
442 most of the authors, deriving ARF from radiative transfer model calculation and  
443 obtaining averages daily values, whose calculations require aerosol information not  
444 known a priori. Thus, in a few cases instantaneous direct measurements of the net fluxes  
445 have been used to derive ARF, and further the effects of atmospheric aerosols on PAR  
446 scarcely have been studied.

447

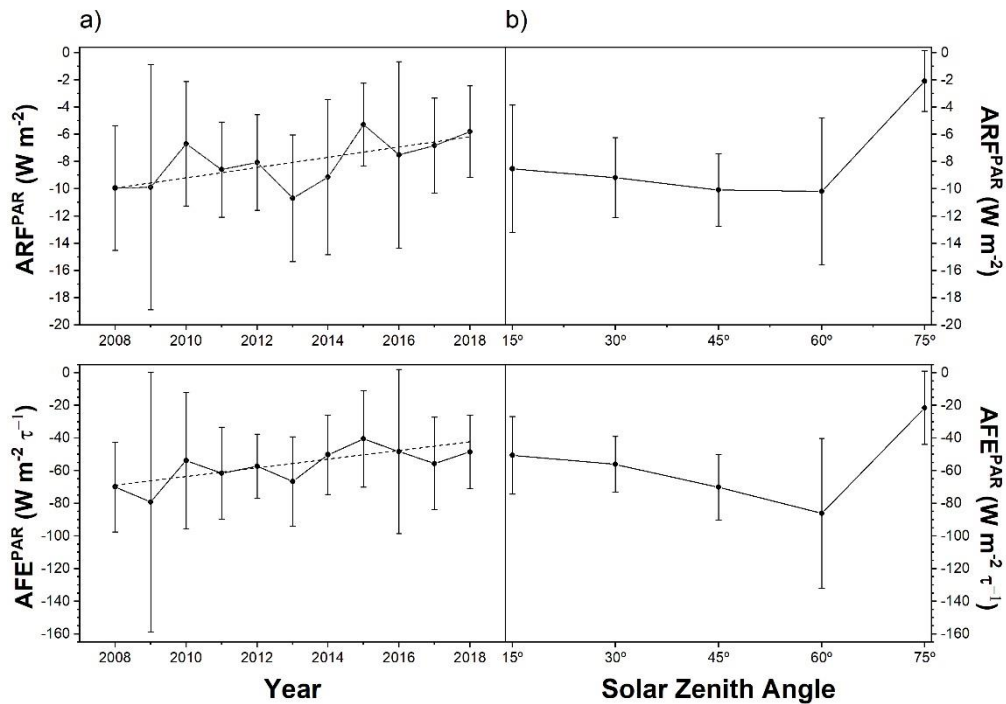
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449



### 450 **4.3 Analysis of AFE and ARF trends**

451 Figures 5b and 6b show the pattern followed for AFE and ARF, with an inflexion point at  
452  $60^\circ$  for both PAR and Total irradiance, showing a similar pattern as described in section  
453 4.2. This similar pattern is explained by the long path at high solar zenith angle, which  
454 includes strong attenuation of direct solar radiation but also more multiple scattering  
455 and hence more scattered light (Lyamani et al., 2006b). The dependence on SZA is more  
456 pronounced for ARF with respect to AFE. In spite of this growth pattern, values of AFE  
457 and ARF are close to be constant for angles smaller than  $45^\circ$  for both Total and PAR, and  
458 this finding are in agreement with previous studies for surface AFE estimated by  
459 radiative transfer model simulations (Formenti et al., 2002; Meloni et al., 2005). On the  
460 other hand, analyzing  $ARF^{PAR}$  and  $AFE^{PAR}$  by years, absolute terms, maximum values have  
461 been found in 2008 and 2009 ( $-10 \text{ Wm}^{-2}$ ,  $-79 \text{ Wm}^{-2}\tau^{-1}$ ), and minimum values of  $-5.3 \text{ Wm}^{-2}$   
462 and  $-41 \text{ Wm}^{-2}\tau^{-1}$  (ARF and AFE, respectively) both in 2015. For Total radiation maximum  
463 values are obtained in 2009 and 2018 ( $-22 \text{ Wm}^{-2}$  and  $-239 \text{ Wm}^{-2}\tau^{-1}$ ), and minimum values  
464 in 2015 ( $-12 \text{ Wm}^{-2}$  and  $-136 \text{ Wm}^{-2}\tau^{-1}$ ) (Figures 5a and 6a).



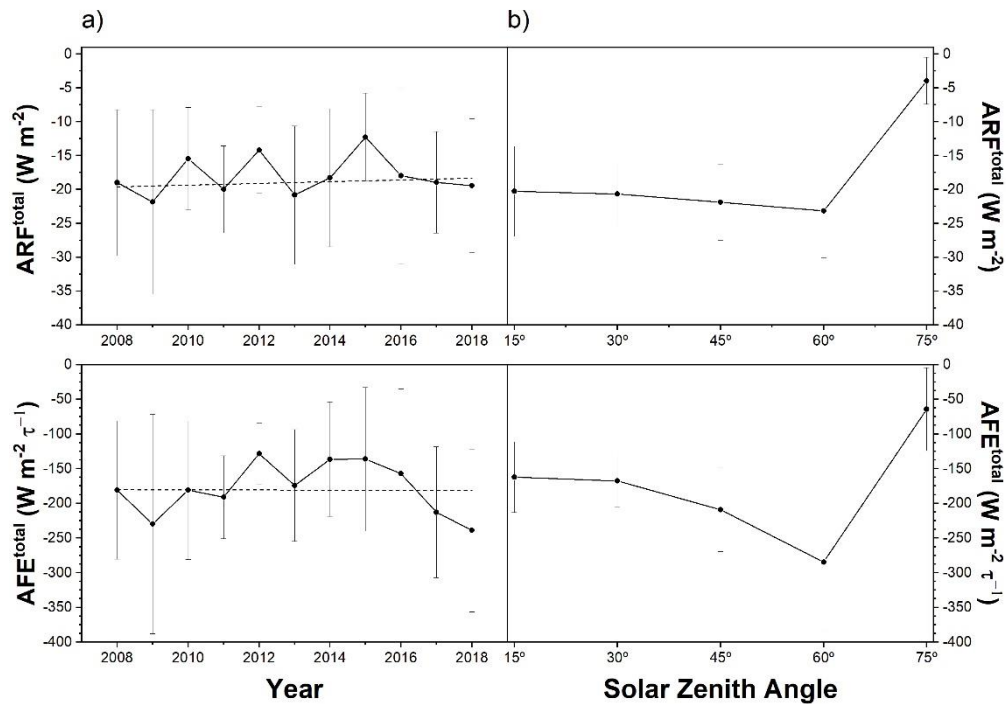
465

466 Figure 5. Evolution of surface aerosol radiative forcing for PAR ( $ARF^{PAR}$ ) and surface aerosol  
 467 forcing efficiency for PAR ( $AFE^{PAR}$ ) with its variability at one standard deviation level by years  
 468 and solar zenith angle (SZA). Dashed lines point out the linear trends evaluated by the Sen  
 469 method.

470

471 The trends analyses revealed for  $ARF^{PAR}$  and  $AFE^{PAR}$  (considering absolute values) a  
 472 downward trend for 2008-2018 period with a slope of  $0.38 \text{ Wm}^{-2}\text{year}^{-1}$  and  $2.66 \text{ Wm}^{-2}\tau^{-1}\text{year}^{-1}$ ,  
 473 respectively, being significant for AFE with a p-value  $< 0.05$  and very close to  
 474 being significant for ARF (p-value = 0.062) (Figure 6a). Although the slope is positive,  
 475 taking into account that the ARF (and AFE) values are negative, the slope value implies  
 476 that the aerosol cooling radiative effect of aerosols is decreasing. The influence of  
 477 calibration factor changes on this trend is negligible based on the long-term stability  
 478 estimated for the PAR sensor along the analyzed period and described in Section 2. This  
 479 result is interesting considering the decrease trend detected for AOD in the last years

480 already commented in section 4.1. However, no upward or downward trend has been  
481 observed for the annual evolution of  $ARF^{total}$  and  $AFE^{total}$  (Figure 6a). In order to consider  
482 a potential compensating effect of other spectral ranges, we have performed the  
483 calculation also with Total minus PAR, i.e. ultraviolet A and B plus near-infrared  
484 irradiance, and no statistically significant trend was found, although the slope is of  
485 opposite sign. This result can be attributed to aerosol properties especially to the  
486 aerosol absorption characteristics. In fact, AFE exhibits a dependence on single  
487 scattering albedo and a larger contribution of the PAR range in relation to Total  
488 irradiance is found for high absorbing aerosols (Mateos et al. 2014). These results  
489 indicate the importance of the knowledge of the PAR, because it is more sensitive  
490 to atmospheric aerosol effects than Total irradiance and, however, it has not been  
491 implemented nowadays at most radiometric stations instruments to measure routinely  
492 the PAR irradiance, unlike Total irradiance, which is a standard variable measured at the  
493 Baseline Surface Radiation Network (BSRN) and many other radiometric stations.



494

495 Figure 6. Evolution of surface aerosol radiative forcing for total irradiance ( $ARF^{total}$ ) and surface  
 496 aerosol forcing efficiency for total irradiance ( $AFE^{total}$ ) with its variability at one standard  
 497 deviation level by years and solar zenith angle (SZA). Dashed lines point out the linear trends  
 498 evaluated by the Sen method.

499

500

## 501 5. CONCLUSIONS

502 Eleven years data set period were analyzed to determine the aerosols radiative effects  
 503 in the photosynthetically active radiation (PAR; 400-700 nm) and Total irradiance (280-  
 504 3000 nm) over an urban site located in a mid-latitude in the Western Mediterranean  
 505 region. These effects have been analyzed through the estimation of aerosol forcing  
 506 efficiency (AFE) and aerosol radiative forcing (ARF), using the direct method, obtaining  
 507 instantaneous values from experimental measurements of aerosol optical depth (AOD)

508 and irradiance measurements. The advantage of this method unlike other methods  
509 using radiative model calculations is that it does not require aerosol information. The  
510 main conclusions are:

- 511 1. A seasonal evolution has been found with maximum AOD<sub>500</sub> values in summer  
512 (0.16 ± 0.09), corresponding to the highest incidence of Saharan dust events and  
513 minimum ones in winter (0.08 ± 0.04). The AFE values ranged between (-12 ± 9)  
514 Wm<sup>-2</sup>τ<sup>-1</sup> and (-198 ± 18) Wm<sup>-2</sup>τ<sup>-1</sup> for PAR while for Total the values varied from (-  
515 9 ± 24) Wm<sup>-2</sup>τ<sup>-1</sup> and (-450 ± 50) Wm<sup>-2</sup>τ<sup>-1</sup>, meanwhile ARF values ranged from (-  
516 1±1) Wm<sup>-2</sup> and (-23 ± 14) Wm<sup>-2</sup> in the case of PAR and from (-1 ± 2) Wm<sup>-2</sup> to (-40 ± 30)  
517 Wm<sup>-2</sup> for Total.
- 518 2. A dependence of both AFE and ARF on solar zenith angle was found with a clear  
519 pattern increasing values of ARF and AFE (in absolute sense) for increasing SZA,  
520 and an inflexion point at 45°-60° range.
- 521 3. The percentage of ARF for PAR with respect to Total irradiance had a mean value  
522 of 50%, a higher value than that obtained for AFE (30%), evidencing the  
523 important impact of atmospheric aerosols on PAR because the ratio PAR/Total  
524 irradiance had a lower average value (43%).
- 525 4. A downward trend for AFE for PAR was found with a slope of 2.7 Wm<sup>-2</sup>τ<sup>-1</sup>year<sup>-1</sup>  
526 with a p-value < 0.05 and no significative trend was found for Total irradiance,  
527 demonstrating that PAR is more sensitive to atmospheric aerosols effects than  
528 Total irradiance and, therefore, evidencing the need to increase the knowledge  
529 of PAR and its interaction with atmospheric aerosols.

530 5. The contribution of different spectral ranges in the trend analysis for AFE can be  
531 governed by the aerosol type, being in general, the visible spectral range the most  
532 dominant, with a variable contribution depending on the aerosol type.

533

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545

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